

Sympathetic Cooling of $^{139}\text{BaH}^+$ Using Laser-Cooled $^{138}\text{Ba}^+$ in Linear RF Traps

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Introduction

Simultaneous precision measurements on both hydrogen and antihydrogen allow the study of the matter-antimatter asymmetry while minimising systematic errors. Examples of such experiments include gravitational tests^[1, 2] and spectroscopy^[3, 4, 5, 6].

The Antihydrogen Laser PHysics Apparatus (ALPHA) at CERN aims to conduct direct precision comparisons between antihydrogen and hydrogen. The first ingredient, antihydrogen, is obtained by mixing a cold positron plasma with a cloud of antiprotons in a Penning-Malmberg trap. This project focuses on investigating the possibility of obtaining the second ingredient, hydrogen, by cooling barium monohydride ions (BaH^+) and subsequently photo-dissociating the ion to create atomic hydrogen with sub-Kelvin temperatures.

We consider obtaining hydrogen as follows. First, $^{138}\text{Ba}^+$ and $^{139}\text{BaH}^+$ ions are laser ablated from the surface of the barium hydride salt target. Subsequently, these ions can be trapped in a linear radio-frequency (RF) trap. Once the charged ions are trapped in electric fields, Ba^+ ions can be laser cooled using the 493 nm transition line. Cool Ba^+ ions then interact with hotter BaH^+ ions via Coulomb forces and the ion ensemble cools down as a whole, which is known as the sympathetic cooling^[7]. Finally, hydrogen is photodissociated from the BaH^+ ions using a ~ 415 nm laser beam and becomes ~ 139 times colder than the ion before photodissociation^[8].

The absence of nuclear spin in $^{138}\text{Ba}^+$ means that there is no hyperfine splitting in electron energy levels. Therefore, these ions are easier to laser cool than e.g. $^{137}\text{Ba}^+$ and $^9\text{Be}^+$.

The use of a linear RF trap has at least two advantages: 1) particles can be introduced or extracted from multiple angles; the lasers can be introduced along at least three different axes; 2) the absence of significant magnetic fields does not cause splitting of otherwise degenerate energy levels which simplifies laser cooling.

In this report we present the results of simulating sympathetic cooling of BaH^+ ions using laser-cooled Ba^+ ions in linear RF quadrupole and octupole traps. We

show that it is possible to bring ions to a crystal state and obtain 0.1 – 100 mK hydrogen. The simulation relied on the ideas used in other RF trap molecular dynamics simulations^[9, 10].

Theory

Linear Quadrupole Trap

A linear RF trap is a device that uses time-varying electric potentials applied on cylindrical rods to trap charged particles. We consider a quadrupole with dimensions and voltages shown in FIG 1 with cylindrical endcaps. As we will later see, this configuration yields a simple ion trajectory stability analysis^[11].

Similar to the original hyperbolic quadrupole trap proposed by Wolfgang Paul^[12], it can be shown that the charges in our trap obey the Mathieu equations,

$$\ddot{x} = (a + 2q \cos 2\tau)x, \quad (1)$$

$$\ddot{y} = (a - 2q \cos 2\tau)y, \quad (2)$$

where $a = \frac{4e\alpha U_{END}}{m\omega_0^2}$ and $q = \frac{2eV_{RF}}{m\omega_0^2}$ are the dimensionless Mathieu parameters, and the derivatives are taken with respect to the dimensionless time $\tau = \frac{1}{2}\omega t$.

Only for certain values of a and q can the ion's trajectory remain bounded. Notice the parameter α in the definition of a . It depends on the geometry of the trap and is due to the assumption that the endcap potential acts on the charged particle radially the same way as middle rods with the potential αU_{end} would and hence can take values in the interval $[-1, 0]$.

For sufficiently high values of ω an adiabatic approximation can be made^[13]. In particular, the radial motion of particles $r(t)$ can be decoupled into micromotion $\delta(t)$ and secular motion $\Delta(t)$: $r(t) = \delta(t) + \Delta(t)$. The former oscillates rapidly with the AC frequency ω of the electrodes with small amplitudes while the latter oscillates slowly with larger amplitudes. Averaging over the micromotion, a harmonic pseudo-potential can be obtained,

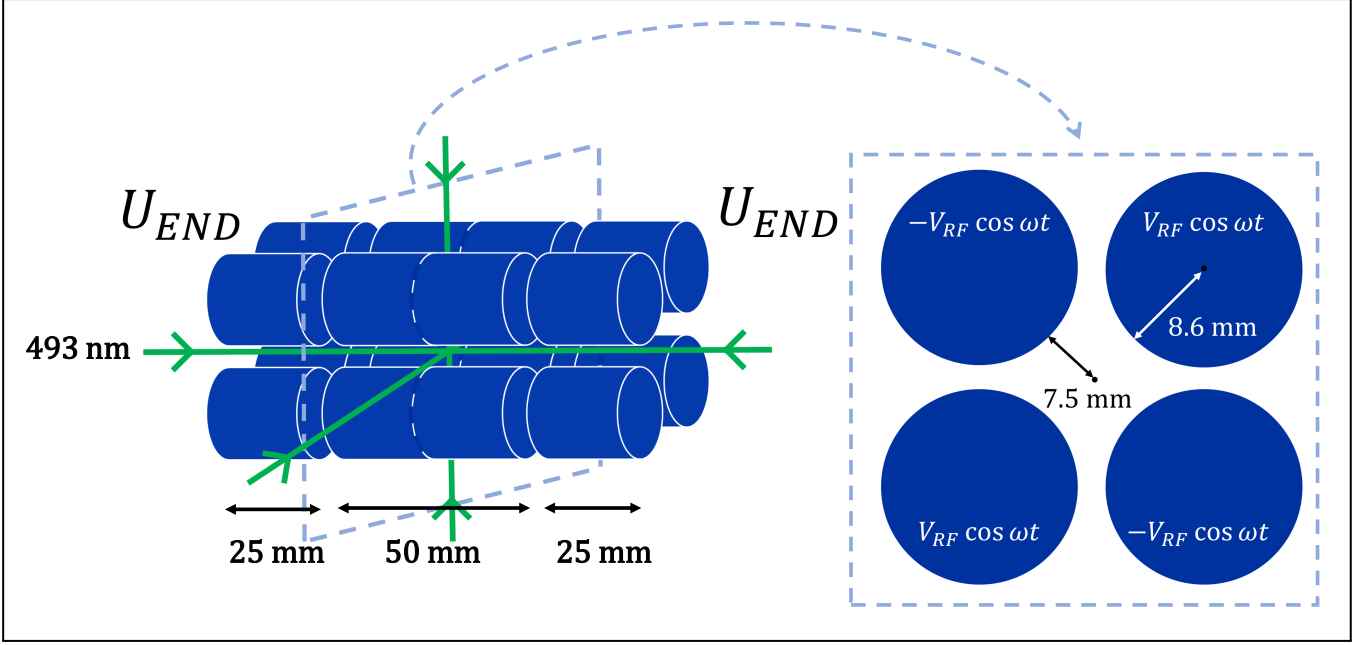


Figure 1. A linear quadrupole RF trap with cylindrical endcaps and its cross-section. A RF-voltage source with amplitude V_{RF} and angular frequency ω is connected to all rods to trap charged particles radially. Additionally, a DC voltage source U_{END} is applied to the endcaps to trap charged particles axially. The distance from the axis to the rod surface is $r_0 = 7.5$ mm and the radius of the rods is 1.1468 times larger. The green arrows indicate the directions of laser-cooling beams with a wavelength 493 nm.

$$V(r, z) = \frac{m\omega^2(a + \frac{q^2}{2})}{8} r^2 - \frac{m\omega^2 a}{4} z^2. \quad (3)$$

The secular motion is due to the force generated by the pseudopotential. Furthermore, it can be shown that the amplitude in micromotion oscillations is proportional to the radial distance from the main axis of the trap, $\delta(t) = \frac{q\Delta(t)}{2} \cos 2\tau$ [13]. Therefore, ions farther away from the trap axis tend to be hotter than the ones closer to the axis.

Linear Octupole Trap

A multipole trap has a flatter potential than a quadrupole trap, and as the number of poles is increased, its potential approximates a box potential. A complex configuration is paid off by the vanishing micromotion in flatter potential regions. Therefore, multipole traps allow to cool ions to lower temperatures than quadrupole traps.

We consider an octupole configuration similar to the quadrupole one but with the reduced rod radius to 2.5 mm as suggested by [14]. The dimensions and voltages are illustrated in FIG 2.

Unfortunately, the stability of ion trajectories in multipole traps depends on the initial position of the ion and hence cannot be described using a two-dimensional

stability parameter space like for the quadrupole trap. However, stability can be described locally using the adiabaticity parameter $\eta = \frac{2e\vec{\nabla} \cdot \vec{E}}{m\omega^2}$, where the ion trajectory is stable for $\eta < 0.3$ [15].

The octupole RF voltage acts on charged particles as follows [16],

$$\ddot{x} = \frac{2ex^3}{mr_0^4} \left(1 - \frac{3y^2}{x^2} \right) V_{RF} \cos \omega t, \quad (4)$$

$$\ddot{y} = \frac{2ey^3}{mr_0^4} \left(1 - \frac{3x^2}{y^2} \right) V_{RF} \cos \omega t. \quad (5)$$

Similar to quadrupoles, an octupole pseudopotential can be derived [14]. As seen in the FIG 3, the RF voltage of the octupole produces a sixth-order radial potential while the DC voltage applied to the endcaps adds a quadratic radial defocussing,

$$V(r, z) = \frac{e^2 V_{RF}}{m\omega^2 r_0^2} \left(\frac{r}{r_0} \right)^6 \quad (6)$$

$$+ \frac{1}{2} e\alpha U_{END} \left(\frac{r}{r_0} \right)^2 - e\alpha U_{END} \left(\frac{z}{r_0} \right)^2. \quad (7)$$

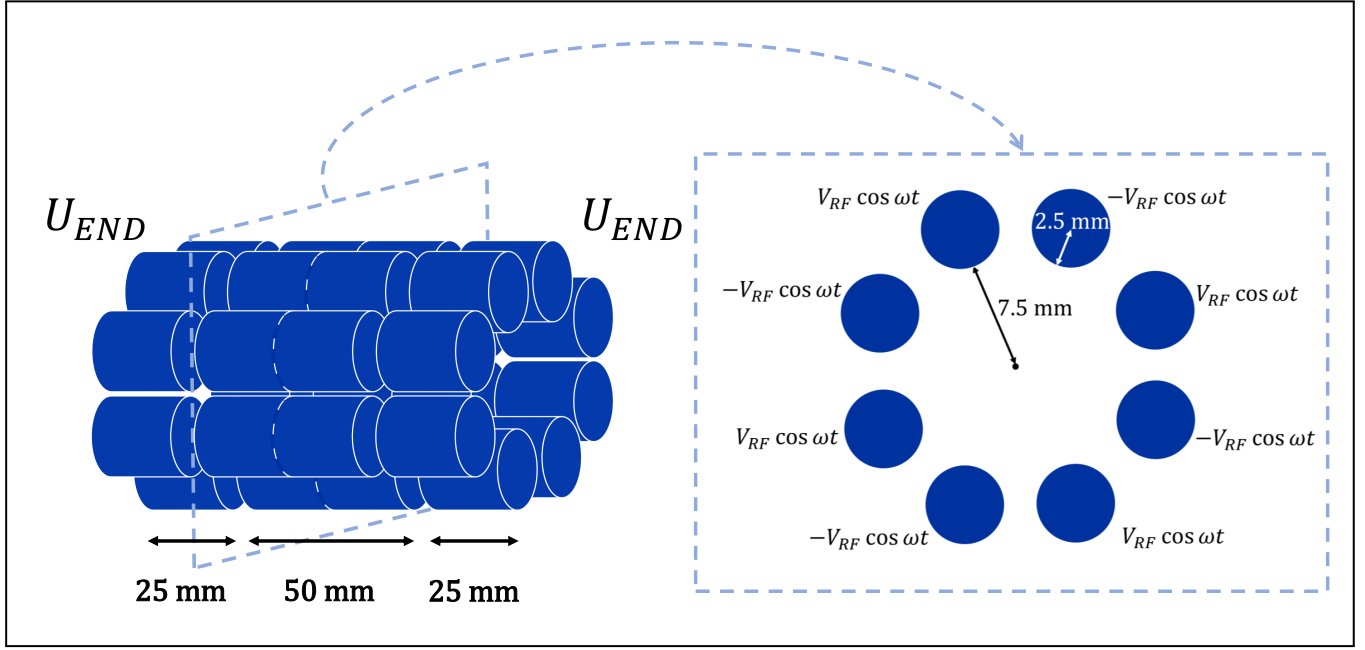


Figure 2. A linear octupole RF trap with cylindrical endcaps and its cross-section. The RF voltages of neighbouring rods have opposite phases.

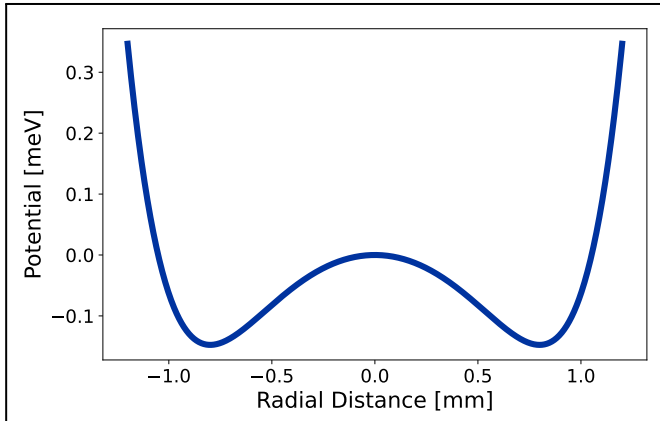


Figure 3. The octupole radial potential is a sum of the non-harmonic RF pseudopotential and a defocussing harmonic endcap potential. The potential minimum is at $r \approx 0.8$ mm.

Laser and Sympathetic Cooling

Assume that barium ions have two electronic energy levels separated by $\Delta E = hf_r$, where h is Planck's constant and f_r is the resonance transition frequency. An incident photon of frequency f gets absorbed by an ion with a probability determined by the Lorentzian probability density function shown in FIG 4. As a result, an electron jumps onto an excited state and sits there before decaying back to the ground state. The decay time follows the exponential distribution with the mean lifetime τ . While decaying, it emits a photon with a frequency following

the same Lorentzian curve.

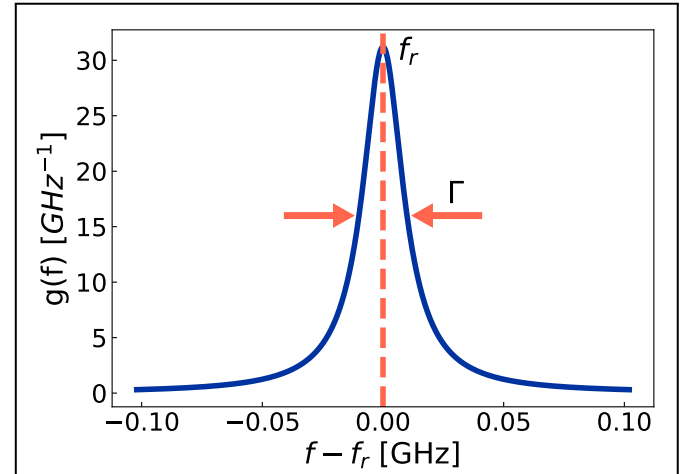


Figure 4. The Lorentzian distribution $g(f)$ which describes the probability of absorbing or emitting a photon with a frequency f .

If the lifetimes were infinite, the Lorentzian would have been a Dirac delta distribution, but due to the Heisenberg uncertainty principle it has a natural linewidth with a full width at half maximum of $\Gamma = \frac{1}{2\pi\tau} \approx 20.4$ MHz.

Now, assume that a barium ion moves with a velocity v projected onto the the laser axis in the opposite direction of the photon propagation. In the frame of the ion, the photon frequency f will be blueshifted to $f' = (1 + \frac{v}{c})f$. Therefore, if the laser frequency is tuned

to a value just below the resonance frequency the ions will only absorb photons moving in the opposite direction of the ion which usually results in slowing down of the ion.

When an ion re-emits a photon, it receives a kick in an arbitrary direction resulting in a random walk in the momentum space. By studying the equilibrium of heating rates due to the emission recoiling and absorption cooling, one can obtain the lowest achievable energy using standard laser cooling called the Doppler limit^[17]. Evaluating the Doppler limit formula for the Ba^+ ions yields $T_{\text{Doppler}} = \frac{h\Gamma}{2k_B} \approx 0.5 \text{ mK}$, where k_B is the Boltzmann constant. Such temperatures are reachable only if the micromotion vanishes and the collisions with nearby particles do not produce heating.

Although BaH^+ ions don't interact with the laser directly, they may get sympathetically cooled^[7] by interacting with the Ba^+ ions via Coulomb interactions, as we will discuss in the next sections.

Methods

Before running a molecular dynamics simulation[†], we needed to pick the trapping parameters. First, we picked the quadrupole stability parameters a and q . To achieve this, we generated stability diagrams using Floquet theory^[18] and a COMSOL Multiphysics simulation (see FIG 5). The COMSOL stability diagram was obtained by releasing a particle with a radial velocity of 100 m/s from the centre of the trap and observing if it escaped the trap during a 10 μs simulation. Initially, the geometric parameter α was set to -1 . Once the stability diagram was obtained, the value of α was found to be -0.024 by stretching the COMSOL-simulated diagram along the a -axis until it matched the Floquet diagram.

For the quadrupole, we picked the voltage amplitudes $U_{\text{END}} = 8 \text{ V}$ and $V_{\text{RF}} = 160 \text{ V}$ with the frequency $f = 500 \text{ kHz}$ satisfying the stability points $(a, q) = (-0.001, 0.4)$. Note that the parameter a was chosen such that $-a < q^2$ to minimise radial defocussing effects.

For the octupole, we tried to pick as large V_{RF} as possible and as small U_{END} as possible to produce flatter trapping radial potential while maintaining stability. We ended up choosing $V_{\text{RF}} = 40 \text{ V}$ and $U_{\text{END}} = 1 \text{ V}$ with the frequency $f = 100 \text{ kHz}$. To verify that these parameters produce a trapping potential with bounded ion trajectories, we performed a COMSOL simulations where we released a $2 \times 2 \times 2 \text{ mm}$ grid of 27 uniformly distributed ions with isotropic velocities 10 m/s from the centre

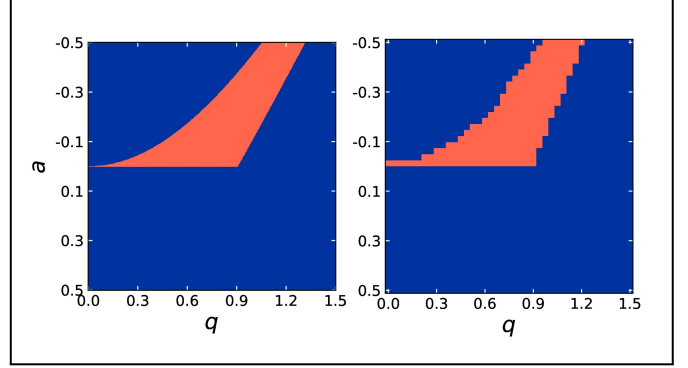


Figure 5. A Floquet stability diagram (left) and a COMSOL stability diagram (right). The (a, q) parameters in the orange region generate a bounded ion trajectory. Note that $a < 0$ to enable axial trapping.

of the trap and observed that all of the ions remained trapped during a 100 μs run.

To determine the octupole geometric parameter $\alpha = -0.039$, we studied the Z-amplitudes of charged particles for a range of velocities in a COMSOL simulation. More specifically, the Z-amplitudes were fitted with a quadratic function Az^2 , as predicted by the equation 7, which yields $\alpha = -\frac{Ar_0^2}{eU_{\text{END}}}$. The resulting radial potential is illustrated in FIG 3.

Once all the necessary parameters were obtained, we wrote a molecular dynamics simulation where the ions were evolved using the Euler method and interacted via Coulomb forces. The timestep of the Euler method was equal to that of the excited state lifetime $\tau = 7.8 \text{ ns}$.

We simulate 500 Ba^+ and 500 BaH^+ ions. At the start of the simulations, the velocities of the ions are isotropic, as illustrated in FIG 6, and follow a Maxwell-Boltzmann distribution at 1 K as shown in FIG 7. Both directions and speeds were selected using the accept-reject algorithm. The initial positions of 1000 ions were uniformly distributed over a cylinder for the quadrupole and a ring for the octupole as depicted in the FIG 8. The dimensions of these shapes were chosen such that they were not too big to avoid particles accelerating due to external electric fields, nor were they too small to avoid a Coulomb explosion where particles gain a significant energy from repulsion due to Coulomb interactions.

As the simulation was set running, at each timestep, the excited ions re-emitted their photons and the ground-state ions could absorb photons which was decided by the accept-reject algorithm following the Lorentzian probability. We used six lasers in total, one counterpropagating pair for each of the X, Y, Z trap axes. The order of lasers used for the accept-reject algorithm was shuffled at random.

[†]The Python codes and COMSOL files can be found on GitHub: https://github.com/NikitaPoljakov/CERN_Summer

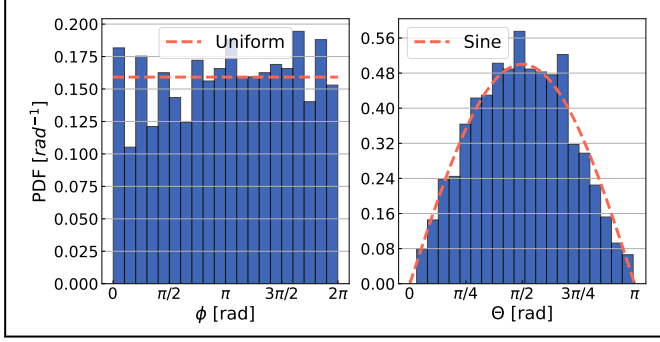


Figure 6. Initial azimuthal and polar velocity directions of 1000 particles. The former follows a uniform distribution and the latter follows a sine distribution such that the initial velocity directions are uniformly distributed over a unit sphere.

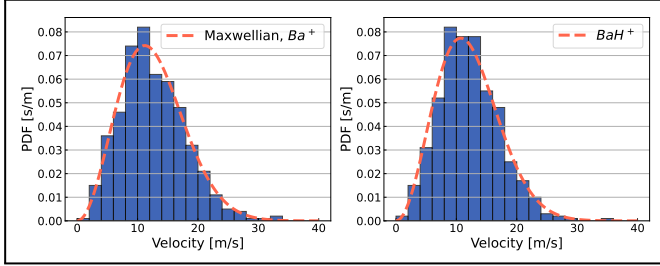


Figure 7. Initial speed distributions of 500 Ba^+ and 500 BaH^+ ions at 1 K following Maxwell-Boltzmann distributions.

Note that we assume sufficiently wide laser beams such that particles at any position in the trap could interact with the laser. For the quadrupole trap, this would yield the beam width radii of $(r_x, r_y, r_z) = (0.1, 0.1, 2)$ [mm], and for the octupole, the beams would be $(r_x, r_y, r_z) = (0.8, 0.8, 2)$ [mm] wide.

To cool down ions at all speeds, the frequency of the laser was swept linearly from the minimal to the maximal value during a 3 ms (sweep rate $\sim \text{GHz/s}$), such that the cooling started with the hotter particles. The starting frequency was equal to the blueshifted resonance frequency by the mean of the initial speed distribution. The final frequency was chosen such that the laser mostly interacted with the particles above the recoil limit^[17] $v_{\text{recoil}} = \frac{hf_r}{2mc}$ below which laser interactions can only cause heating.

Results and Discussion

We present results for three cases: 1) laser-cooled ions in the RF regime; 2) laser-cooled ions in the pseudopotential regime; 3) non-cooled ions in the RF regime. We focus on the first case as it is the closest approximation to the hypothetical experiment and include the results for the

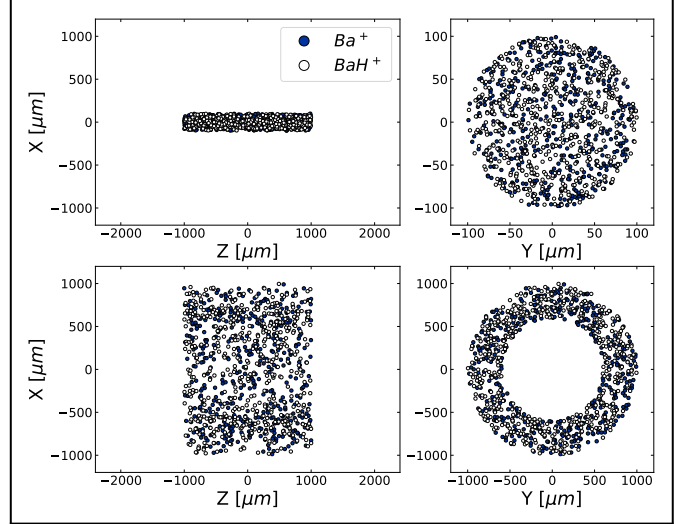


Figure 8. Initial positions of 500 Ba^+ and 500 BaH^+ ions for the quadrupole (top) and an octupole (bottom) in the XZ (left) and XY (right) planes. For the quadrupole case, the ions are uniformly distributed over a cylinder of radius 0.1 mm and height of 2 mm. For the octupole case, the ions are uniformly distributed over a ring of radius 0.8 mm, radial thickness 0.4 mm and axial length 2 mm.

latter two cases for comparison. The final positions of ions for the three cases are depicted in FIG 9, 10, and 11, respectively.

For the laser-cooled cases we see that the quadrupole ions group into ellipsoidal shapes. Note that only in the pseudopotential case the ions follow radial symmetry. The symmetry breaking in the RF case might be explained by the initial voltage phases in the simulation. Furthermore, we can see the ions forming two denser radial layers, one at the boundary and one at the half-radius of the radial cross-section.

In the octupole simulations, the ions form a ring along the radial potential minimum. Similar to the quadrupole case, the ring is not perfectly circular in the RF case but follows a fourfold radial symmetry, while the pseudopotential simulation generates a ring with a circular boundary. Moreover, in the pseudopotential case the octupole ions group into circular z-layers.

In the simulation without the laser-cooling, ions group into similar shapes as in the other cases but with more dispersed positions.

Returning to the simulation with the RF-forces and laser-cooling, we take a radial cross-section of the quadrupole ion ensemble, as illustrated in FIG 12. We find that there is a clear boundary and internal structure indicating that the ions are laser-cooled to a crystal state. Although the octupole ion ring in FIG 9

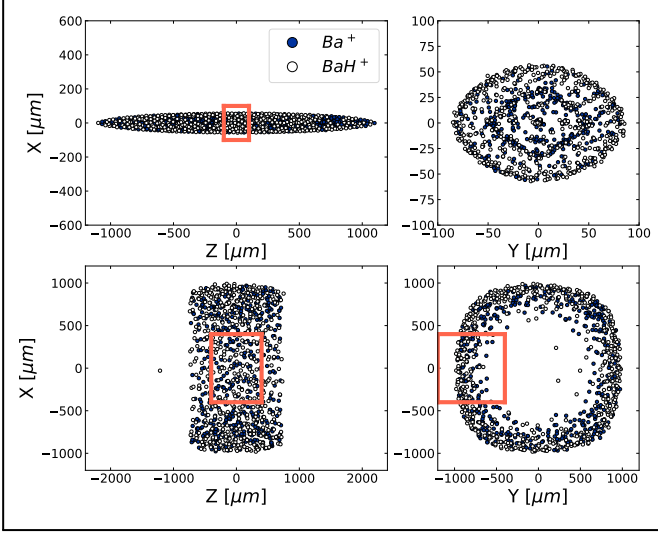


Figure 9. Final positions of 500 Ba^+ and 500 BaH^+ ions subject to quadrupole (top) and octupole (bottom) RF-forces after laser cooling. The regions marked with orange boundaries are shown in FIG 12 and 13.

has a clear outermost boundary, it does not have an innermost one. This might be because the radial potential is steeper in the outermost part of the ring. Furthermore, a zoomed-in picture in FIG 13 does not indicate any internal structure and hence the laser-cooled octupole ions are in a liquid-gas state.

In FIG 12, we see that for the quadrupole case the ions form a Coulomb crystal which is periodic in the Z -direction with the lattice parameter $\sim 35 \mu\text{m}$. The lighter Ba^+ ions are slightly closer to the axis than the heavier BaH^+ ions. Similarly, as we zoom into the octupole ion ring in FIG 13 we see that there are more Ba^+ ions near the radial centre of the trap. Such separation occurs because the total energy of the ensemble is minimized if the lighter species occupy the space of lower potential^[10].

To quantify how hot the final state of ions is, we have accumulated the ion velocities during the last secular period of the simulation in FIG 14. Due to micromotion the ensemble is not colder than its 1 K initial state. In FIG 15, we can see that in the absence of micromotion, the ions can be cooled to sub-mK temperatures. Returning to the RF-simulations, we get a speed cut-off at $\sim 45 \text{ m/s}$ for the quadrupole and the cut-off is at $\sim 32 \text{ m/s}$ for the octupole. Note that the ions do not follow the Maxwell-Boltzmann distribution due to the combination of lasers and RF-forces. This claim is supported by the speed distributions obtained from the two simulations, one with pseudoforces and with laser-cooling, another with RF-forces and without laser-cooling, where the final ion speeds in both simulations approximately follow the Maxwell-Boltzmann distributions as seen in FIG 15 and 16.

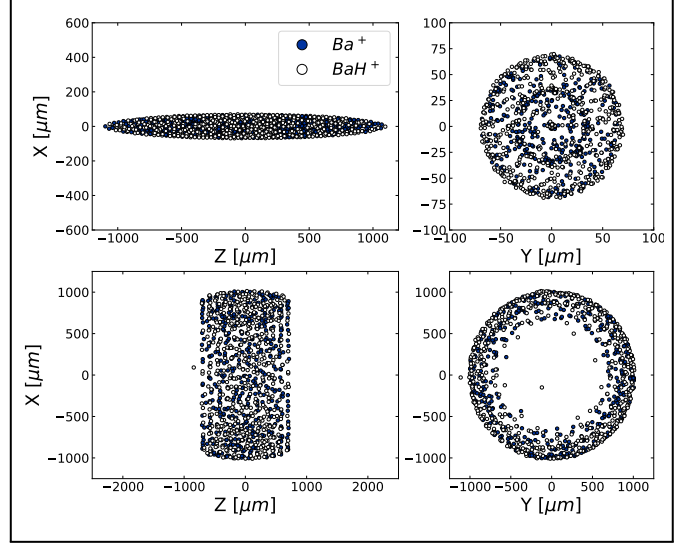


Figure 10. Final positions of 500 Ba^+ and 500 BaH^+ ions evolved in quadrupole and octupole pseudoforces with laser-cooling.

Notice that without lasers, the quadrupole ions heat up more than the octupole ions. This might be either due to the steeper quadrupole potential gradient which generates more micromotion or it may be due to the initial positions of ions being significantly further from the equilibrium positions than in the octupole case.

Coming back to the simulation with the RF-forces and laser-cooling, in FIG 17 we show the immediate temperatures of the hydrogen atoms if we photodissociated all BaH^+ ions at once. We have assumed that the energy carried by a dissociating photon is equal to the bonding energy between Ba^+ and H and hence the daughter hydrogen atom moves with the same speed as its parent BaH^+ ion. We then converted the BaH^+ velocity distributions into temperature distributions using $\frac{3}{2}k_B T = \frac{1}{2}m_H v_{\text{BaH}}^2$, where we have assumed the equipartition theorem. Note that this assumption could have been more accurate if we left the ions reach thermal equilibrium after turning off the lasers.

For the quadrupole, approximately 37%, 12%, 5% of hydrogen atoms reach temperatures below 10, 1, 0.1 mK, respectively. The average quadrupole hydrogen temperature is 25 mK. For the octupole, the percentages are approximately 53%, 15%, 3%, respectively. The average octupole hydrogen temperature is 12 mK. Therefore, although there are slightly more ultracold hydrogen atoms in the quadrupole trap, the octupole allows to reach a lower average temperature. This might be due to a flatter potential minimum where micromotion is less significant than in the quadrupole case.

A typical minimum B-filed Ioffe-Pritchard trap used for trapping antihydrogen in ALPHA is $\sim 0.5 \text{ K}$ meaning

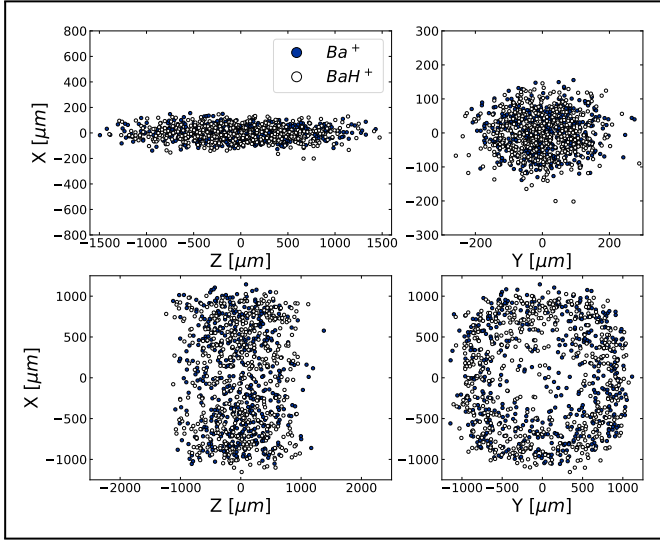


Figure 11. Final positions of 500 Ba^+ and 500 BaH^+ ions evolved in quadrupole and octupole RF-forces without laser-cooling.

that all of the hydrogen produced in our simulations can foreseeably be trapped for precision measurements in the same ALPHA apparatus where antihydrogen precision measurements take place^[19].

Notice that in a quadrupole trap ions get hotter with the radial distance as seen in FIG 18. This temperature gradient can be used to achieve even lower hydrogen temperatures. Instead of photodissociating BaH^+ ions over the whole ensemble, we can perform this only with the ions close to the axis. In particular, by shining a photodissociating laser beam of radius $5\text{ }\mu\text{m}$ along the trap z -axis, we would obtain $\sim 50\text{ }\mu\text{K}$ hydrogen. However, such approach would produce less hydrogen unless it is possible to cool down new BaH^+ ions from the outermost layers of the ensemble and replace the dissociated Ba^+ ions near the axis.

In future investigations, finite cooling laser beam widths could be included in the simulation. Furthermore, the behaviour with higher initial temperatures could be studied and the temperature evolution of ions investigated. To make the simulation more realistic, trap imperfections, such as the residual gas and rod asymmetries could be incorporated.

Alongside laser and sympathetic cooling, other cooling methods can be studied. For example, adiabatic cooling^[20] can be used to bring ion ensembles to even lower temperatures by lowering the pseudopotential and making ions perform work on the potential walls. However, this method will cause ions to occupy larger space and fewer ions will have negligible micromotion.

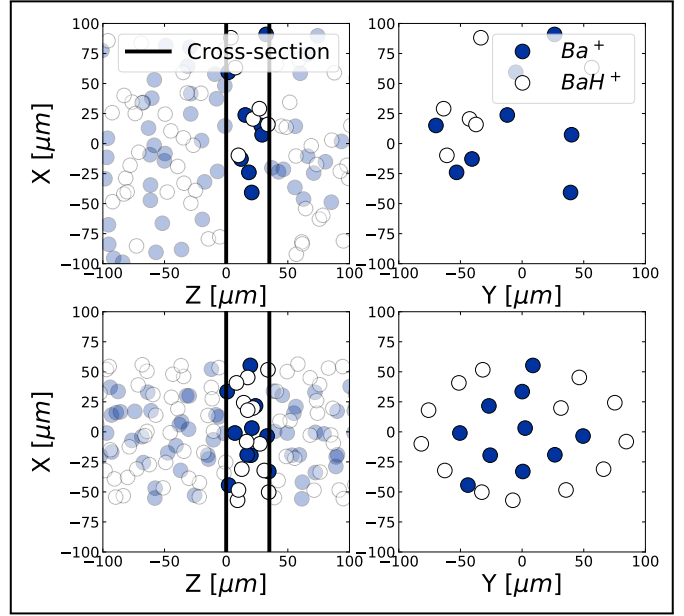


Figure 12. Cross-sections of the initial (top) and final (bottom) positions of ions in the $z \in [0\text{ }\mu\text{m}, 35\text{ }\mu\text{m}]$ axial interval in the quadrupole simulation with RF-forces and laser-cooling.

Conclusion

Trapping and cooling of $^{138}\text{Ba}^+$ and $^{139}\text{BaH}^+$ ions has been simulated in linear RF quadrupole and octupole traps. We considered two specific quadrupole and octupole configurations and showed that after photodissociation of BaH^+ ions, hydrogen reaches lower temperatures in the octupole case with the mean value of 11 mK than in the quadrupole case with the mean value of 24 mK .

Acknowledgments

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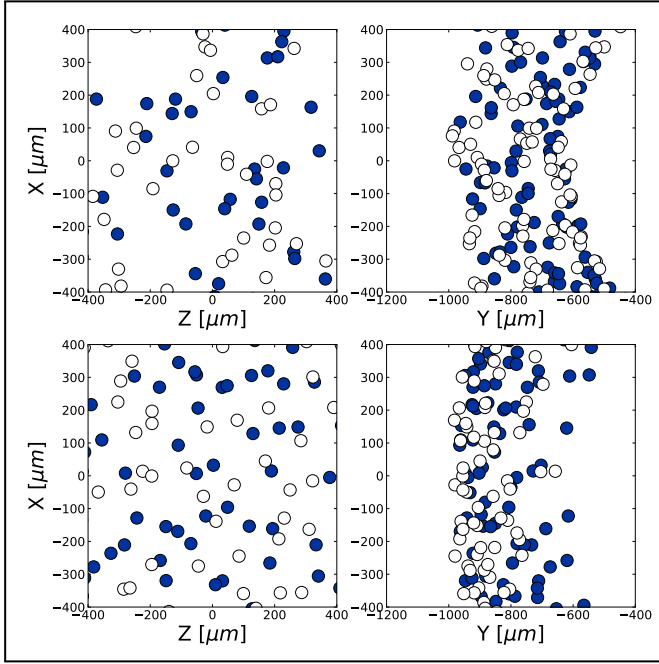


Figure 13. Zoomed-in plots of the initial (top) and final (bottom) ion positions in the octupole simulation subject to RF-forces and laser-cooling.

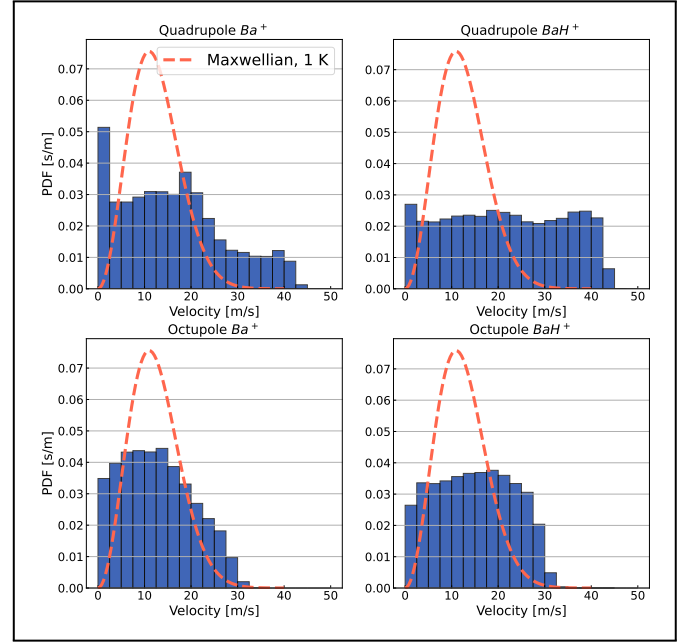


Figure 14. Speed distributions accumulated over the final secular period of the simulation for the with RF-forces and laser-cooling. The accumulated speeds are compared to the initial 1 K Maxwell-Boltzmann distribution.

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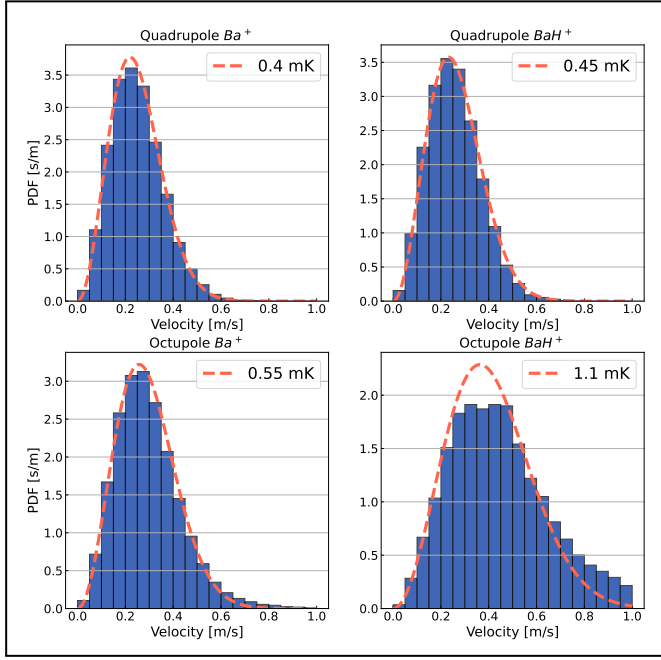


Figure 15. Speed distributions accumulated over the final secular period for simulation with pseudoforces and laser-cooling. Every speed histogram approximates Maxwell-Boltzmann distribution.

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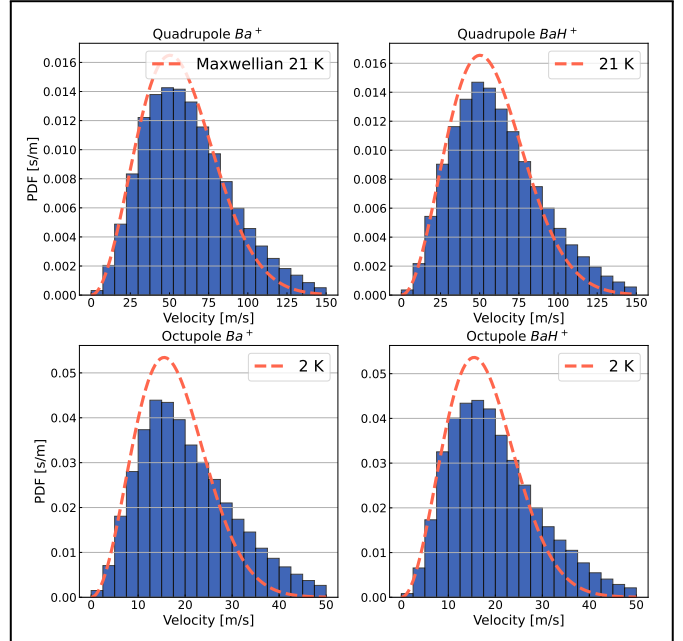


Figure 16. Speed distributions accumulated over the final secular period for simulations with RF-forces and without laser-cooling. Every speed histogram approximates Maxwell-Boltzmann distribution with a deviating tail which might be due to hotter particles experiencing larger potential and micromotion.

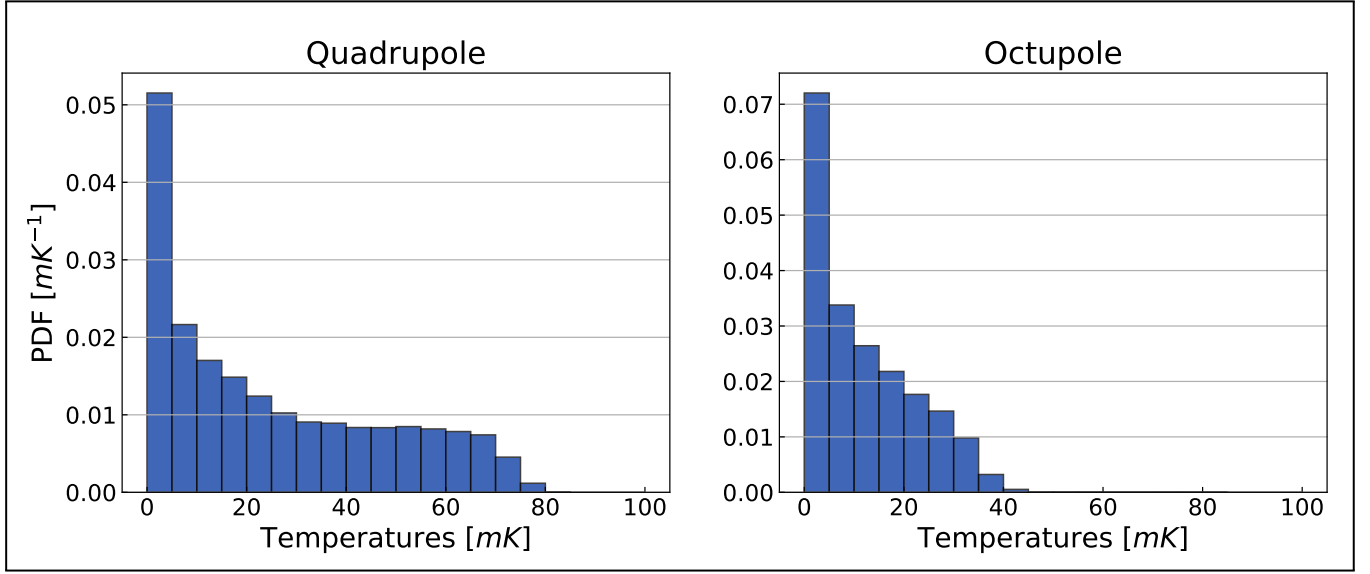


Figure 17. Hydrogen temperature distribution after photodissociating all BaH^+ ions at once at the end of the simulation with RF-forces and laser-cooling.

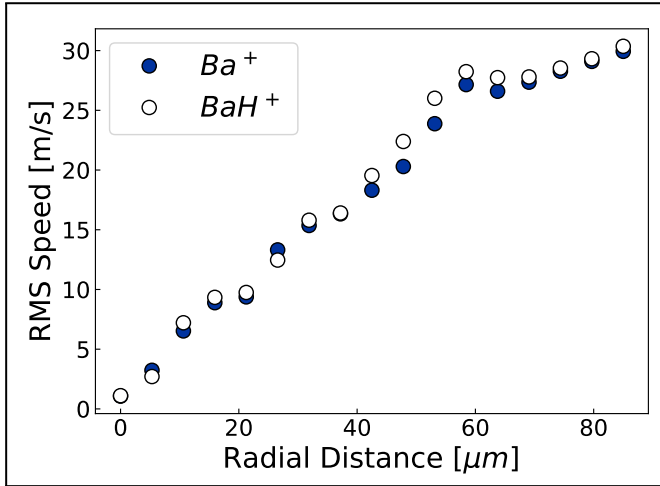


Figure 18. RMS speeds of ions belonging to $5 \mu\text{m}$ thick radial layers in the equilibrium ellipsoidal shape produced in the quadrupole simulation with RF-forces and laser-cooling. As expected, the micromotion increases with the radial distance. BaH^+ ions in the first shell have a RMS speed of $\sim 1.1 \text{ m/s}$.